

# NonOhmic Conduction as a Probe of Coexisting Phases in Manganites

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A new scaling formalism is used to analyze nonlinear  $I$ - $V$  data in the vicinity of metal-insulator transitions (MIT) in five manganite systems. An exponent, called nonlinearity exponent, and an onset field for nonlinearity, both characteristic of the system under study, are obtained from the analysis. The onset field is found to have an anomalously low value corroborating the theoretically predicted electronically soft phases. The scaling functions above and below the MIT of a polycrystalline sample are found to be same but with different exponents which are attributed to the distribution of the MIT temperatures. The existence of field scales in manganites underlines the universality of the assumed scaling form in disordered systems.

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In an intermediate doped manganite  $A_xB_{1-x}MnO_3$  ( $A$ = trivalent rare-earth and  $B$ = divalent alkaline-earth), a metal-insulator transition (MIT) usually manifests itself in the form of a resistance peak at a temperature  $T_{MI}$  close to the Curie temperature  $T_c$  and is accompanied by colossal magnetoresistance. The resistance curve is rather asymmetric with a sharp drop on the lower side of  $T_{MI}$ , especially in single crystals. Spatial microscopy experiments[1, 2] and theories[3, 4] attribute this to nanoscale phase separation near the transition where the spin-sensitive transport occurs through a percolative network of ferromagnetic metallic (FM) and paramagnetic insulating (PI) domains. Features such as formation of a backbone of FM domains[1, 5] at  $T_{MI}$ , significant increase in noise upon approaching  $T_{MI}$ [6] indeed conform qualitatively to the standard percolation model[7]. However, the nature of the MIT which is first-order and large deviations in transport exponents in manganites from those in standard percolation model indicate that the present understanding of the coexistent phases is still incomplete[8].

The intrinsic inhomogeneity due to phase separation creates conditions favorable for nonlinear (or nonOhmic) transport due to wide variations of local electric fields inside a sample. Study of field dependent transport in manganites through the phase separated regions can be used as a tool to probe further complexities in conduction mechanisms beyond those revealed in zero field measurements. Nonlinear conduction has been earlier studied in manganites mostly in charge ordered (CO) regimes[9], in materials without CO[10], and in orbital ordered samples[11]. Analysis of the nonlinear data so far has been devoid of any systematics and mostly empirical.

Nonlinear studies[12] in composites of metals and insulators at room temperature, which are supposed to be prototypes of the percolation model, reveal that above the percolation threshold (below  $T_{MI}$  in the present case) and for not too large bias, the (electric) field-dependent conductivity  $\sigma(p, F)$  of a sample is given by the model-independent

scaling relation

$$\frac{\sigma(p, F)}{\sigma(p, 0)} = \Phi\left(\frac{F}{F_o}\right). \quad (1)$$

Here  $\Phi$  is a scaling function,  $p$  is the metal fraction and  $F$  is the applied dc electric field. The field scale  $F_o(p)$  is given by[13]

$$F_o(p) \sim \sigma_o^{x_p}, \quad (2)$$

where  $\sigma_o(p) = \sigma(p, 0)$  is the zero bias conductivity at a metal fraction  $p$ . The nonlinearity exponent  $x_p$  in 3D systems was found to have experimental values of about 0.45[12, 13]. The latter is consistent with a model prediction  $x_p \leq \nu/t$ . Here,  $\nu$  and  $t$  are correlation and conductivity exponents[7] respectively. In contrast, literature below the percolation threshold (above  $T_{MI}$ ) is rather scant in this case. Eq. 1 describes a situation where nonlinearity is investigated as a function of disorder (i.e.  $p$ ). However, as seen below, nonlinearity can also be investigated as a function of temperature, for example, at a fixed or varying disorder with a possibility of different exponent  $x_T$ . Recently, a similar scaling framework[14] has been proposed for disordered systems with localized states. In manganites, disorder, aside from spin disorder, arises from random potential fluctuations due to  $A^{3+}$  and  $B^{2+}$  ion cores, Jahn-Teller distortion and local trapping in ferromagnetic regions with non-collinear magnetic order[15]. Manganites thus provide unique opportunity to study the applicability of such a scaling formalism.

In this letter, we present evidence that states around metal-insulator transition peaks indeed obey the scaling (Eq. 1) in both single crystal and polycrystalline samples of manganites. This proves that origin of nonlinearity is intrinsic, and not due to extraneous factors such as inelastic intergrain tunneling. The scaling functions on both sides of a MIT in a polycrystalline sample are found to be identical albeit with different nonlinearity exponents  $x_T$ . The electric field scale  $F_o$  is anomalously found to be of the order of 1-10 V/cm, about three orders of magnitude less

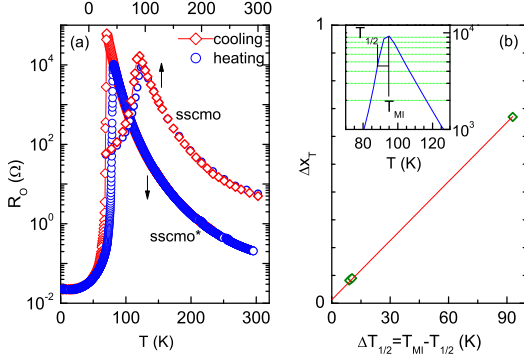


FIG. 1. (Color online) a) Ohmic resistance  $R_0$  vs. temperature  $T$  of two SSCMO samples - one single crystal (with star) and another polycrystalline. The symbols, circle and diamond, correspond to the data of heating and cooling cycles respectively; b) Plot of  $\Delta x_T$  vs.  $\Delta T_{1/2}$  for four polycrystalline samples (Table I). The inset illustrates using the SSCMO data in the panel a how  $\Delta T_{1/2}$  is defined. See text for further explanation.

than that in granular composites, corroborating the claim of electronically soft phases in manganites[16]. In contrast to those in granular materials, nonlinearity exponents in manganites often seem to possess negative signs.

An important factor in choosing samples for the present study (Table I) was the requirement to have resistance changes between the baseline and the peak on either side of a MIT as large as possible to extract reliably the nonlinearity exponent (see Eq. 2). Accordingly, systems with Sm were chosen[17].  $\text{La}_{0.275}\text{Pr}_{0.35}\text{Ca}_{0.375}\text{MnO}_3$  was chosen for its robust phase separation property[1, 6]. A single crystal of  $\text{Sm}_{0.55}(\text{Sr}_{0.5}\text{Ca}_{0.5})_{0.45}\text{MnO}_3$  (SSCMO\* in Table I) was grown using the floating zone technique[18]. Four different polycrystalline samples (ones in Table I without star sign) were prepared by the usual solid-state reactions. Two SSCMO samples have slightly different chemical composition. Of all the samples, LMO is self-doped[19] while rest are the usual hole-doped. The values of  $T_{MI}$  for different samples analyzed in this work are shown in Table I.  $I$ - $V$  measurements were done in a Janis cryotip with bar-shaped samples placed on sapphire substrates. Data were taken under constant current condition. Maximum current levels were kept low to minimize Joule heating in the samples. Both four probe and two probe contacts gave the same results. All measurements reported here were done at zero magnetic field.

Figure 1 shows resistance-temperature ( $R$ - $T$ ) plots for two SSCMO samples - one single crystal (with star) and another polycrystalline. The single crystal (SC) shows a first-order-like sharp MIT transition for  $T \leq T_{MI}$  whereas the transition to FM state in the polycrystalline (PC) sample is rather gradual. Such behavior, widely known in literatures, results from a distribution of  $T_{MI}$ [20]. The hysteresis between cooling and warming cycles is also illustrative of first-order phase transition. Steady state  $I$ - $V$  measure-

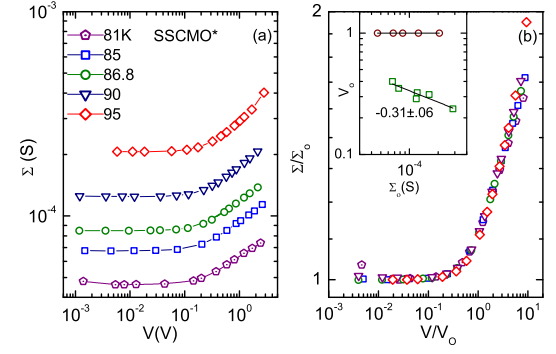


FIG. 2. (Color online) (a) Conductance vs. bias in the insulating paramagnetic (PI) phase of a SSCMO single crystal at different temperatures ( $T > T_{MI}$ ) as indicated. (b) Scaling of the data in the panel a. Inset shows bias scale,  $V_0$  vs. linear conductance,  $\Sigma_0$  for two experimental paths. See text for details. The number represents the slope of the linear fit to the data.

ments at  $T \leq T_{MI}$  are not feasible in the SC sample as the sharp transition is metastable with huge relaxation times ( $> 5$  hours). The same problem arises in PC samples during cooling cycle but is absent in the heating cycle. This makes PC samples a sort of necessity in the present study if measurements are to be carried out on both sides of the MIT. The  $\Sigma - V$  ( $\Sigma = 1/R$ ) characteristics of SC in the warming cycle at different temperatures ranging from 85 to 105 K ( $T > T_{MI}$ ) are plotted on a log-log scale in the left panel of Fig. 2(a). The nonlinear response of conductance to the application of bias  $V$  is clearly seen in the figure. The onset bias  $V_0(T)$  is the bias at which conductance starts deviating from its linear value  $\Sigma_0(T)$  at temperature  $T$  and is defined (arbitrarily) such that  $\Sigma(V_0) \approx 1.1\Sigma_0$ . The plot of the normalized conductance  $\Sigma(T, V)/\Sigma(T, 0)$  vs. the normalized bias  $V/V_0$  using the data of the PI phase of Fig. 2a is shown in Fig. 2b which exhibits almost perfect data collapse, thus validating Eq. 1. The bias scale  $V_0$  appears to be independent of temperature and is shown in the inset of Fig. 2b (circles) as a function of the Ohmic conductance. The nonlinearity exponent  $x_T^{PI}$  is thus zero in the PI phase of the SSCMO single crystal. The experimental path in this case corresponded to cooling the sample down to a temperature far below  $T_{MI}$  and then gradual heating. To check the effect of possible hysteresis, the sample in another experiment was first cooled from the room temperature to 85 K slightly above  $T_{MI}$  (i.e. still in the PI phase) where heating and cooling cycle coincided (Fig. 1), and then heated. The exponent  $x_T^{PI}$  in this case turns out to be  $-0.31 \pm 0.06$  (squares in the inset of Fig. 2b). Such sensitivity of the exponent to the experimental paths perhaps confirm the role of hysteresis.

The fraction of the FM phase for  $T > T_{MI}$  is supposed to increase as  $T$  is decreased[1, 20]. Such a situation is comparable to a non-percolating granular samples with different  $p$ . We have analyzed such data at a

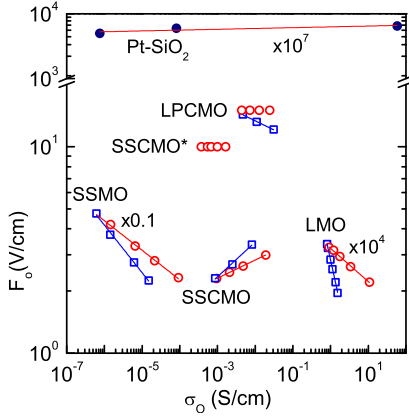


FIG. 3. (Color online) Plots of onset field  $F_o$  vs. Ohmic conductivity  $\sigma_o$  for different systems as marked. Circles and squares correspond to PI and FM phases respectively. Data of LMO and Pt-SiO<sub>2</sub> were shifted to right by the factors as mentioned.

fixed temperature of three samples of Pt-SiO<sub>2</sub> with different metal fractions[21] (supplementary materials). Interestingly, the nonlinearity exponent  $x_p$  in this case is found to be  $\approx 0$  as in SSCMO\*. It is seen however from Table I that the same exponent  $x_T^{PI}$  in the polycrystalline samples can have non-zero values, both positive and negative although the magnitudes are not large. The field scale  $F_o$  in the prevalent theories[21, 22] is given by the condition that the field-activated conduction becomes comparable to the temperature-activated one i.e.,

$$eF_o w = k_B T. \quad (3)$$

Here  $w$ , a relevant length scale, may be the mean separation length between two metallic clusters in a granular system [21] or the average hopping distance in a variable range hopping system[22]. If  $w$  and consequently,  $F_o$  remain independent of  $p$  we have  $x_p = 0$ . This means that in a manganite system even though amount of the FM phase may vary with  $T$ , the average tunneling distance between two FM clusters in PI phase remains fixed. This however can not explain any non-zero exponent. Let us now consider another aspect of the field scale, namely its magnitude which is displayed in Fig. 3 for both SSCMO\* and Pt-SiO<sub>2</sub>. It is seen that the scale in SSCMO\* is about three orders of magnitude less than that in Pt-SiO<sub>2</sub>. This can be easily understood by noting that a carrier moves from one metallic cluster to another which are chemically and structurally same but distinguished only by spin or charge ordering. Hence, the energy barrier that a carrier must overcome must be much less than that in a medium of chemically dissimilar materials like Pt and SiO<sub>2</sub>. On the other hand, taking the separation distance in a manganite as large as 100 nm[1] a field of 10 V/cm ( $F_o$  in SSCMO\*, Fig. 3) corresponds to an anomalously low barrier height of about 1 K compared to about 300 K estimated from noise studies[23]. Furthermore, such low value of 1 K contradicts the basis of

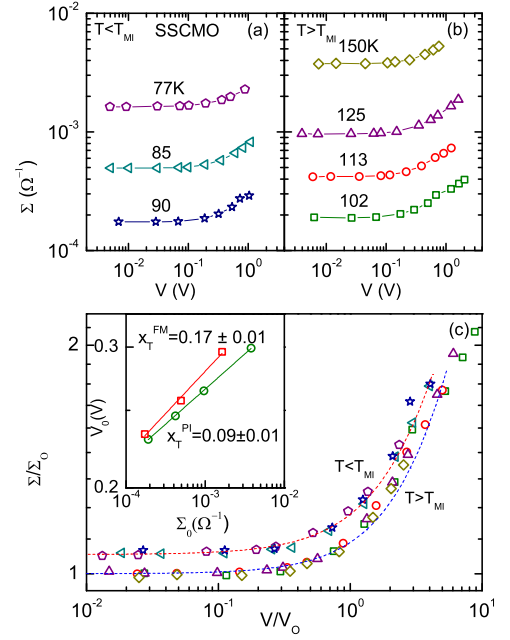


FIG. 4. (Color online) The upper panels show plots of conductance vs. bias in a SSCMO polycrystalline sample in the ferromagnetic metallic phase (FM) for  $T < T_{MI}$  (a) and PI phase for  $T > T_{MI}$  (b) at temperatures as indicated. The lower panel (c) shows the data collapse of the same data of the upper panels. The scaled curve of the FM side is shifted for clarity. Inset shows two log-log plots of  $F_o$  vs.  $\sigma_o$  data corresponding to PI (open circle) and FM sides (open square) respectively. The solid lines are linear fits to the data with slopes  $x_T^{PI}$  and  $x_T^{FM}$  respectively.

nonlinearity in Eq. 3, the temperature range of interest being 81-95 K. It may be noted that Eq. 3 is not compatible with Eq. 2 which is a typical power-law relation in critical phenomena.

The nonlinear  $I$ - $V$  curves could also be caused by Joule heating. A simple mean field approach shows that  $V_o \sim \Sigma_0^{-0.5}$  or,  $x^{Joule} \approx -0.5$ [24]. As none of the exponents obtained is close to this value it can be concluded that the nonOhmic behavior is an intrinsic feature and not caused by heating. Let us now turn to measurements on a host of polycrystalline samples (Table I). Since results in various samples are basically similar, we consider only the results for the polycrystalline SSCMO in Fig. 4 for the purpose of comparing results from the single crystal of the same system discussed above.  $\Sigma - V$  plots at various temperatures both below and above  $T_{MI}$  are shown in Figs. 4a and 4b respectively. Note that in both cases conductance increases from bottom to top. Curves in panels a and b look qualitatively similar but exhibit a subtle difference as shown below. Both sets of data were found to follow the same scaling as in Eq. 1 and actually collapse on the same curve (Fig. 4c). But the scaled data in FM phase ( $T < T_{MI}$ ) has been shifted in the figure for the sake of clarity.  $V_o$  thus obtained are shown in log-log plots (open squares for FM

TABLE I. Sample parameters and nonlinearity exponents. See text for definitions.

System	Abbreviation	Type	$T_{MI}$ K	$T_{1/2}$ K	$x_T^{FM}$	$x_T^{PI}$
$\text{Sm}_{0.55}(\text{Sr}_{0.5}\text{Ca}_{0.5})_{0.45}\text{MnO}_3$	SSCMO*	Single crystal	82	-	-	0
$\text{Sm}_{0.55}\text{Sr}_{0.3375}\text{Ca}_{0.1125}\text{MnO}_3$	SSCMO	Polycrystal	94	85	$0.17 \pm 0.01$	$0.09 \pm 0.01$
$\text{Sm}_{0.55}\text{Sr}_{0.45}\text{MnO}_3$	SSMO	Polycrystal	69.5	59.7	$-0.23 \pm 0.01$	$-0.14 \pm 0.01$
$\text{La}_{0.275}\text{Pr}_{0.35}\text{Ca}_{0.375}\text{MnO}_3$	LPCMO	Polycrystal	113	102.6	$-0.14 \pm 0.02$	0
$\text{La}_{0.87}(\text{Mn}_2\text{O}_3)_{0.13}\text{MnO}_3$	LMO	Polycrystal	155	62	$-0.83 \pm 0.01$	$-0.16 \pm 0.01$

phase and open circles for PI phase) in the inset of Fig. 4c. The solid lines are power-law fits to Eq. 2 with the exponents as indicated. The exponents in two sides of the MIT are different, but are positive and non-zero in contrast to zero value in the single crystal. Moreover,  $V_o$  is generally less in polycrystals than in the single crystal (Fig. 3). This is contrary to what will be expected if the origin of nonlinearity were due to intergrain effects. It is seen from Table I that  $|x_T^{PI}|$  is generally less than  $|x_T^{FM}|$ .  $F_o$  as a function of conductivity for all polycrystalline samples are shown in Fig. 3.

The FM phase at  $T < T_{MI}$  possesses a continuous metallic path or a backbone through a sample. Conductivity of a backbone is too large to make any contribution to nonlinearity. The nonlinear conduction above the threshold in standard percolation is thought to arise from tunneling across thinly separated conducting bonds dangling off the backbone[12], in contrast to tunneling of a carrier from one metallic cluster to another across insulating barrier below the threshold. Thus, the observation of the same scaling function above and below  $T_{MI}$  in the polycrystalline samples was unexpected. Alternatively, as mentioned earlier, there exists a distribution of  $T_{MI}$  and hence, a fraction of PI clusters which gets progressively reduced as temperature is decreased [20]. This is indirectly supported by the fact that the sign as well as the magnitude of the exponents  $x_T^{FM}$  are generally very different from  $\sim 0.45$  found in the binary composites above the threshold. Conduction across such PI clusters may give rise to same scaling functions on both sides of  $T_{MI}$ . It is then necessary to explain why the exponents on both sides of the transition are different. We postulate that the difference is related to the width of the distribution of  $T_{MI}$ , which is taken to be Gaussian[20]. A measure of the width, denoted by  $\Delta T_{1/2}$ , is assumed in this work to be the difference between  $T_{MI}$  and the temperature  $T_{1/2}$  such that  $R_o(T_{1/2}) = R_o(T_{MI})/2$  (see inset of Fig.1b). The absolute difference in exponents,  $\Delta x_T = |x_T^{PI} - x_T^{FM}|$  when plotted against  $\Delta T_{1/2}$  yielded a straight line passing nearly through the origin (Fig. 1b). Of course, no explanation of the individual exponents, particularly negative ones, is available at present.

In summary, we suggested a model-independent scaling formalism to analyze the electric field-dependent conduction data in manganites. It was shown how quantities such as the field scale or nonlinearity exponents could be used

to gather further information about the underlying physical states. There is a need for theoretical modeling to explain great variety of exponents found in this work. The same scaling analysis could be applied to other regimes such as charge ordered states in manganites.

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